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Thermal-strain-induced enhancement of electromagnetic properties of SiC–MgB_2 composites

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The effect of thermal strain caused by the different thermal expansion coefficients (α) of the MgB_2 and SiC phases on the electromagnetic properties was studied for SiC–MgB_2 composite, which was made by premixing SiC and B, followed by Mg diffusion and reaction. Thermal strain in the MgB_2 phase was demonstrated with x-ray diffraction, Raman spectroscopy, and transmission electron microscopy. In contrast to the common practice of improving the critical current density J_c and the upper critical field H_{c2} of MgB_2 through chemical substitution, by taking advantage of residual thermal strains, we are able to design a composite showing only a small decrease in the critical temperature and a little increase in resistivity but a significant improvement over the J_c and H_{c2} of pure MgB_2. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3078396]

Strain engineering has been used to modify the properties of materials in ferroelectric,[1] superconducting,[2] and ferromagnetic thin films.[3] Here, we report the observation of residual thermal stress/strain in dense SiC–MgB_2 superconductor composites prepared by a diffusion method. The doping of nano-SiC particles into MgB_2 has been proven to be particularly effective in significantly enhancing the critical current density J_c, the irreversibility field H_{irr}, and the upper critical field H_{c2}.[4–8] Extensive work has been carried out to understand the mechanism behind the special doping effects of C and SiC.[4,5,9] Recently, a dual reaction model has been proposed to provide a more comprehensive understanding of nano-SiC doping in MgB_2.[5] The disadvantages of nano-SiC doping are the reduction in the critical temperature T_c and the increase in resistivity, resulting in the deterioration of low-field J_c.

In this work, we designed and fabricated a SiC–MgB_2 composite by a diffusion reaction of Mg with a premixed B–SiC bulk. Starting powders of crystalline B (99.999%), with and without 10 wt % SiC particles, were mixed and pressed into pellets. The pellets were then put into an iron tube filled with Mg powder (99.8%). The atomic ratio between Mg and B was 1.15:2.0. Since the formation of MgB_2 was accomplished via a diffusion process, long reaction times were thus needed to obtain a fully reacted MgB_2 bulk. The samples were sintered at 923–1223 K for 10 h in a quartz tube, where a flow of high purity argon gas was maintained, and then furnace-cooled to room temperature. All samples were characterized by x-ray diffraction (XRD) and analyzed using Rietveld refinement. Microstructural observations were performed by using scanning electron microscopy (SEM), field emission SEM, and transmission electron microscopy (TEM). The magnetization of samples was measured at 5 and 20 K using a quantum design physical properties measurement system. The irreversibility field H_{irr} and H_{c2} could be deduced using the criteria of 0.1 and 0.9 of $\rho(H,T)$, respectively.

The *a*- and *c*-axis lattice parameters and the SiC content were determined as shown in Table I. We note that the *a*-axis parameter is virtually the same for both the doped and undoped samples, while the *c*-axis parameter is slightly enlarged in the SiC–MgB_2 composite. In contrast, the *a*-axis parameter for *in situ* processed SiC doped MgB_2 is reduced, while the *c*-axis parameter generally remains unchanged, as reported by a number of groups.[4,5] It is also interesting to note that the SiC particles remained unreacted and formed a composite with the MgB_2 in the SiC-doped sample. The Rietveld refinement analysis results showed that there was about 9.3 wt % SiC, which is about the same as in the starting precursor. This result may be due to both the crystalline, rather than amorphous, nature of the SiC and the diffusion process and is consistent with the fact that the XRD patterns showed no presence of Mg_2Si, in clear contrast to SiC-doped MgB_2 prepared by the *in situ* technique.[4,5] Mg_2Si is always present in such *in situ* samples due to the reaction of Mg with SiC. The reason why the SiC additives did not decompose and react with the Mg is largely attributed to the fact that crystalline SiC was used, which is not as reactive as amorphous SiC, as has been reported previously. Crystalline SiC has been used as a substrate material, and it showed complete compatibility with MgB_2 films with no evidence of any reaction.[9]

There is a small drop (0.6 K) in T_c and a small increase in ρ(40 K) (from 12 to 16 $\mu\Omega$ cm) for the SiC-doped sample. In contrast, *in situ* processed SiC doped MgB_2 normally experiences a decrease in T_c from 1.5 to 2 K (Ref. 9) and an increase in ρ(40 K) from 90 to 300 $\mu\Omega$ cm.[5] These SiC–MgB_2 composite samples had a high T_c value (37.8 K) and low ρ(40 K) (Table I). It is interesting to note that the SiC–MgB_2 composite sample showed not only an improved in-field J_c but also a lack of degradation and even an enhancement in the self-field J_c, as shown in Fig. 1(a) and Table I, while it has been well established that nano-SiC doping using the conventional *in situ* technique reduces the

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TABLE I. Summary of physical properties of pure and SiC–MgB_2 composite samples.

Sample	Density (g/cm^3)	*a*- and *c*-axis parameters (Å)	Defect induced nonuniform lattice strain (XRD) *a* -axis (%)	Defect induced nonuniform lattice strain (XRD) *c* -axis (%)	Mg vacancies XRD (%)	Grain size TEM (nm)	T_c (K)	ρ(40 K) ($\mu\Omega$ cm)
Pure	1.86	3.085 and 3.5230	0.208	0.292	3.2	~100	38.4	12
10% SiC	1.91	3.084 and 3.5282	0.306	1.13	3.6	~100	37.8	16

self-field J_c.[1] The H_{irr} and H_{c2} of the SiC–MgB_2 sample are significantly improved in comparison with the pure sample, as shown in Fig. 1(b).

The above results raise a serious question concerning the nature of the mechanism behind the significant property enhancement in SiC–MgB_2 composite, as these effects cannot be explained by the dual reaction model[4] or C substitution.[4,5] Because the residual SiC is the dominating impurity, this enabled us to isolate the special effects of residual SiC. As the difference in the thermal expansion coefficients (α) between SiC and MgB_2 is quite large, which can create thermal strains in the MgB_2 matrix during cooling, we can examine whether the residual thermal strain could be an effective pinning mechanism.

The difference in α between MgB_2 and SiC dictates the stress status of the MgB_2 and causes thermal strain in the MgB_2 matrix around the SiC particles during the cooling process. The lattice changes and lattice strain in the MgB_2 matrix during cooling can be derived from the α data.[10,11] If the two phases are strongly bonded, the MgB_2 phase will be subjected to a tensile strain on cooling. In particular, there will be a larger strain along the *c*-axis since there is a larger difference in α along the *c*-axis between MgB_2 and SiC. Our calculation indicated that the lattice strain is as large as -0.55% in the MgB_2 along the *c*-axis at room temperature. This value is approximately equal to the critical strain rate measured for multifilament MgB_2 wires,[12] which showed that the J_c increased with increasing tensile strain rate, although this strain was measured on the macroscale. The increased strain in the SiC–MgB_2 is also evident from the larger full width at half maximum (FWHM) values [0.48° for the (002) peak] than for pure MgB_2 [0.326° for the (002) peak]. The increase in the FWHM can be solely attributed to the increase in the strain since the grain sizes of both samples are the same. Using Williamson–Hall analysis, we calculated the lattice strain to be 0.208% and 0.306% along the *a*-axis and 0.292% and 1.13% along the *c*-axis for the pure and SiC–MgB_2 samples, respectively (Table I).

The thermal strain could create a huge stress field, structural defects, and lattice distortion in the MgB_2 matrix, which would remain in the SiC–MgB_2 sample during cooling. TEM examination revealed the following features. (1) SiC and MgB_2 remain as separate phases and form a strongly bonded composite, as shown in Figs. 2(a) and 2(b). The SiC particles, while micron sized in scale, are comprised of smaller grains a few nanometers in size. It is these small grains that assist in providing excellent wetting and contact with the MgB_2 phase. (2) The grain size for both pure MgB_2 and MgB_2 in the SiC–MgB_2 composite is the same (~100 nm). (3) There is a high density of defects (dislocations and lattice distortion) in the MgB_2 phase along the interfaces between the SiC and MgB_2, as indicated by the arrows in Fig. 2(a), which may be attributable to the tensile strain in the MgB_2 phase that is imposed by the small thermal expansion of SiC during cooling. Figure 2(b) shows the interface between the two coexisting phases of MgB_2 and SiC. The electron diffraction patterns taken from both sides indicate that the two phases are faceted with the [101] plane of SiC and the [001] plane of MgB_2. The thermal expansion coefficient for MgB_2 is highly anisotropic with a large variation in the [001] direction, while that for SiC is nearly isotropic. Thus, this kind of interface will impose tensile stress along the *c*-axis in MgB_2. In comparison, these wave structures are hardly seen in the pure MgB_2 sample. Careful examination indicated that these fringes were induced by lattice mismatch between two layers, so dislocations and lattice distortions were commonly observed in the areas showing the fringes. The lattice distortion observed in TEM confirmed

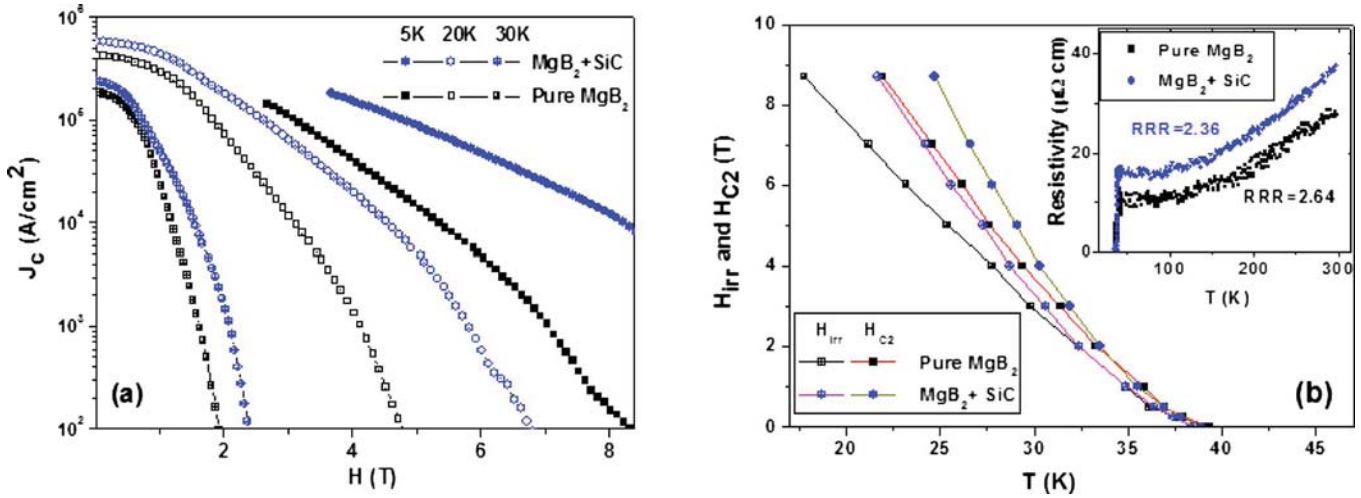

FIG. 1. (Color online) (a) The magnetic J_c vs field at 5 and 20 K for the pure and the nano-SiC doped samples and (b) upper critical field (H_{c2}) and irreversibility field (H_{irr}) as functions of the temperature. The inset shows the resistivity of these samples as a function of temperature.

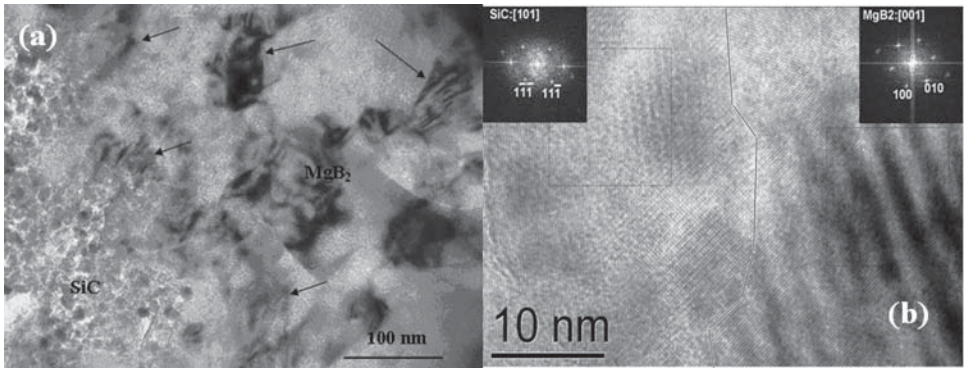

FIG. 2. Bright field TEM images of the two-phase composite and defects along the boundaries between the SiC particles and the MgB_2 matrix. Defects are indicated by arrows, and magnification is indicated by the length of the respective scale bar, which is (a) 100 nm, (b) 10 nm. (b) shows a high resolution TEM image of the interface with inset electron diffraction patterns of SiC and MgB_2 from each side of the interface. The middle line shows the actual interface of SiC and MgB_2, which indicates the strong bonding of MgB_2 with SiC.

our Raman measurement results, which are described below.

Raman spectroscopy is known to be an excellent probe for the detection and estimation of these stresses and strains or lattice distortions. The peaks centered at the phonon density of states (PDOS) are understood to arise due to disorder and distortion.[13] A compressive stress is indicated by a shift of the Raman peak to higher frequencies, while a tensile stress is indicated by a shift of the Raman peak to lower frequencies.[14] The relative intensity and FWHM of the PDOS peaks reflect the extent of the distortion.[15] Figure 3 shows the normalized ambient Raman spectra of the samples sintered at 1123 K for 10 h. The E_{2g} and PDOS peaks are shifted to lower frequency for the SiC-doped sample (E_{2g}:576 cm^{-1}, PDOS:762 cm^{-1}) compared with the pure sample (E_{2g}:600 cm^{-1}, PDOS:770 cm^{-1}), which indicates that there is residual tensile strain in the original $SiC-MgB_2$ composite sample. By comparing the Raman spectra of samples before and after cooling to low temperature (down to 10 K), we find that the most obvious changes in the Raman peaks are that the shifting tendency of the two main Raman peaks, the FWHM of both peaks, and the intensity of the main PDOS peak are increased by the cooling process for the SiC-doped sample but not changed for the pure sample.

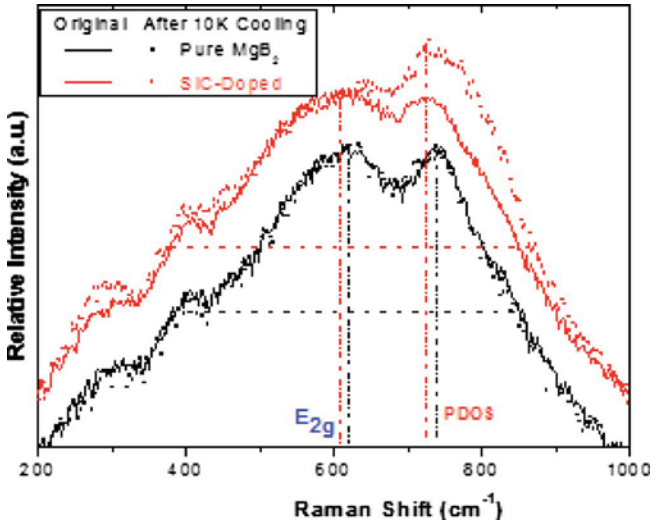

FIG. 3. (Color online) The normalized ambient Raman spectra of samples sintered at 850 °C for 10 h. The line spectra correspond to measurements taken before, and the dot spectra to measurements taken after cooling to 10 K for pure MgB_2 (black) and $SiC+MgB_2$ (red) composite.

The enhanced PDOS peak in the SiC-doped sample is an indication of the increased residual strain, which leads to enhanced flux pinning and thus improves the J_c and H_{c2}.

The present findings have a significant implication, as they open up a new direction for numerous bulk composites that can be strain-engineered to achieve desirable material properties without significant alteration in intrinsic properties, as compared to chemical substitution.

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[1]K. J. Choi, M. Biegalski, Y. L. Li, A. Sharan, J. Schubert, R. Uecker, P. Reiche, Y. B. Chen, X. Q. Pan, V. Gopalan, L. Q. Chen, D. G. Schlom, and C. B. Eom, Science **306**, 1005 (2004).
[2]I. Bozovic, G. Logvenov, I. Belca, B. Narimbetov, and I. Sveklo, Phys. Rev. Lett. **89**, 107001 (2002).
[3]S. K. Streiffer, J. A. Eastman, D. D. Fong, C. Thompson, A. Munkholm, M. V. R. Murty, O. Auciello, G. R. Bai, and G. B. Stephenson, Phys. Rev. Lett. **89**, 067601 (2002).
[4]S. X. Dou, O. Shcherbakova, W. K. Yoeh, J. H. Kim, S. Soltanian, X. L. Wang, C. Senatore, R. Flukiger, M. Dhalle, O. Husnjak, and E. Babic, Phys. Rev. Lett. **98**, 097002 (2007).
[5]A. Matsumoto, H. Kumakura, H. Kitaguchi, B. J. Senkowicz, M. C. Jewell, E. E. Hellstrom, Y. Zhu, P. M. Voyles, and D. C. Larbalestier, Appl. Phys. Lett. **89**, 132508 (2006).
[6]M. D. Sumption, M. Bhatia, M. Rindfleisch, M. Tomsic, S. Soltanian, S. X. Dou, and E. W. Collings, Appl. Phys. Lett. **86**, 092507 (2005).
[7]Y. Zhu, A. Matsumoto, B. J. Senkowicz, H. Kumakura, H. Kitaguchi, M. C. Jewell, E. E. Hellstrom, D. C. Larbalestier, and P. M. Voyles, J. Appl. Phys. **102**, 013913 (2007).
[8]K. Togano, T. Nakane, H. Fujii, H. Takeya, and H. Kumakura, Supercond. Sci. Technol. **19**, L17 (2006).
[9]X. H. Zeng, A. V. Pogrebnyakov, M. H. Zhu, J. E. Jones, X. X. Xi, S. Y. Xu, E. Wertz, Q. Li, J. M. Redwing, J. Lettieri, V. Vaithyanathan, D. G. Schlom, Z. K. Liu, O. Trithaveesak, and J. Schubert, Appl. Phys. Lett. **82**, 2097 (2003).
[10]J. J. Neumeier, T. Tomita, M. Debessai, J. S. Schilling, P. W. Barnes, D. G. Hinks, J. D. Jorgensen, Phys. Rev. B **72**, 220505 (2005).
[11]J. D. Jorgensen, D. G. Hinks, and S. Short, Phys. Rev. B **63**, 224522 (2001).
[12]M. Dhalle, H. van Weeren, S. Wessel, A. den Ouden, H. H. J. ten Kate, I. Husek, P. Kovac, S. Schlachter, and W. Goldacker, Supercond. Sci. Technol. **18**, S253 (2005).
[13]K. P. Bohnen, R. Heid, and B. Renker, Phys. Rev. Lett. **86**, 5771 (2001).
[14]V. Paillard, P. Puech, R. Sirvin, S. Hamma, and P. R. I. Cabarrocas, J. Appl. Phys. **90**, 3276 (2001).
[15]W. X. Li, Y. Li, R. H. Chen, R. Zeng, S. X. Dou, M. Y. Zhu, and H. M. Jin, Phys. Rev. B **77**, 094517 (2008).